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U-235 GAMMA RADIATION

by

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INTRODUCTION

The three naturally occurring uranium isotopes, U-234, U-235 and U-238, are spontaneous alpha emitters. In a previous report [1] it was shown that the U-234 alpha decay is accompanied by x-rays, and the present paper reports the discovery and measurement of gamma rays accompanying the alpha decay of U-235. The 162 Kev gamma ray found associated with the U-235 decay was discovered and clearly identified at this laboratory in April, 1946. A similar radiation was observed somewhat earlier at the University of Chicago, but was attributed to U-234.

Discovery of the Gamma Radiation

Sample Preparation. The significant steps in the purification of the samples of U-235 used were as follows. The bulk of the radioactive and other impurities were removed by ether extraction of uranyl nitrate from a nitric acid solution of the original ore. Reduction and precipitation of the uranium as UF_4 probably removed few impurities other than the very volatile fluorides such as silicon tetrafluoride. The precipitated, dried UF_4 was then converted with gaseous fluorine to the volatile UF_6, which was purified by condensation and re-evaporation. The UF_6 gas was then passed through a great many fine pored filters which increased by isotopic separation the proportion of U-235 to about one-third, and the UF_6 was then condensed directly into a clean uncontaminated nickel cylinder with quarter-inch walls. It is felt that no radioactive materials except U-234, U-235, U-238 and insignificant traces of radon were present at the time of condensation into the sample cylinder.

Radiation. A large sample of solid U-235 enriched UF_6 in a quarter-inch wall nickel cylinder was placed at a standard distance (about six inches) from a screen wall Geiger counter. An initial activity of about 1000 c/m was observed. Superimposed on this was a growing activity of 24 day half-life (UX_1, UX_2 and UZ), extrapolating to 300-400 c/m at equilibrium. To prove that a residue of the non-volatile fluoride of UX_1, possibly swept through the filters and condensed with the sample, was not the cause of the initial 1000 c/m activity, a sample of natural uranium was examined. This was prepared in the same manner as the preceding sample but was passed through only a single filter with somewhat larger pores. The initial gamma activity of this sample was only slightly above background (net rate 24 c/m) and increased with a 24 day half-life toward 500 c/m. Since this sample contained half again as much U-238, parent of the UX_1, UX_2 and UZ, as the enriched sample, the equilibrium activity of the latter sample should have been about 330 c/m. The initial activity found then (100 c/m) was clearly not attributable to UX_1 inclusion in the sample of enriched uranium to the extent

1. Macklin, R. L. and Knight, G. B., "X-Rays Associated With U-234", Phys. Rev. 72, 435 (1947).

of more than a few percent. Indeed, the initial activity (24 c/m) of the natural uranium sample with much poorer filtration led to the expectation that UX_1 dust carryover in the enriched UF_6 sample was entirely negligible.

Energy of the Radiation

A gram sample of nearly pure U-235 F_4 was used for preliminary absorption measurements. These showed a prominent gamma ray component at about 162 Kev.

For more accurate measurements, UF_6 samples such as those described above were hydrolysed and heated to give U_3O_8 powder. This was spread on sample holders in a thin layer of measured mass. Samples of enriched uranium (one-third U-235, two-thirds U-238) were prepared at the same time and made up with equal quantities of U-238. Thus, the gamma and beta rays of UX_1, UX_2 and UX at any time were equal for a pair of samples. Hence, the difference in counting rate of a pair corresponded to a known quantity of U-235 (or U-234). Absorption measurements with both lead and aluminum were made at these laboratories and later (July, 1946) at Clinton National Laboratories. The 162 Kev gamma radiation was confirmed (see Fig. 1) and two weaker radiations corresponding to L and M X-rays were found. These latter were later shown to be associated with U-234 (see reference 1).

The yield of gamma rays per alpha disintegration can be only roughly estimated. Values near 100% of the U-235 alpha disintegration rate seem most reasonable although the gamma counter efficiencies are so poorly known as to make this little more than a guess. One type counter yielded estimates of 10-100% and another gave 125 - 250%.

Origin of the Radiation

To determine which uranium isotope gave rise to the gamma activity observed, samples of varying composition were used. The experiments with natural and enriched uranium already described showed, of course, that the 162 Kev gamma activity was not proportional to the U-238 content. Two samples were obtained which were nearly equal in U-234 content but differed by a factor of three in U-235 content as shown by mass spectrometer. These samples were several months old and hence contained virtually equilibrium quantities of UX_1, UX_2, and UZ proportional to the U-238 present.

Using the previous results for the gamma radiation relative intensities measured through a quarter-inch of nickel, the ratio of gamma activities of these samples (through 1/4" of nickel) was predicted on the basis of two hypotheses. If the 162 Kev gamma activity were due to the U-234, the activity ratio should be 0.65 because of the difference in U-238, UX_1, UX_2 and UZ content of the two samples. However, if the 162 Kev gamma activity were due to the U-235, the ratio would be 2.32. The ratio observed was 2.31 ± 0.04. Hence, the gamma ray appears definitely to be associated with U-235.

Discussion

The high U-234 alpha activity of available U-235 samples has made it impossible to observe alpha gamma coincidences or to discover U-235 alpha rays unaccompanied by gamma rays. The latter alpha rays would be expected to differ in energy from those of U-234 by only 40 Kev. Alpha ray spectrographs capable of resolving these alpha ray groups require a far higher specific activity than uranium possesses. Hence, experimental confirmation of the decay scheme proposed is not at present available.

The alpha decay of U-235 is supposed to lead in most, perhaps all, cases to an excited state of Th-231 (UY) which rapidly emits a 162 Kev gamma ray to reach the ground state. A half-life for deexcitation longer than ten minutes would have been readily observed. The conversion of the gamma ray has not been detected and is probably slight.

The decay of UY (Th-231) from its ground state has been investigated and reported separately[2].

Summary

Evidence for the existence of a gamma ray accompanying the alpha decay of U-235 is presented. The energy of the gamma ray, as determined by several independent absorption measurements, is 162 Kev.

(Note: Figure 1 is Drawing #LDA-303 and identical with Figure 1 of the earlier Report K-97)

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2. Knight, G. B. and R. L. Macklin "Radiations of UY", Phys. Rev. 75, 34 (1949).

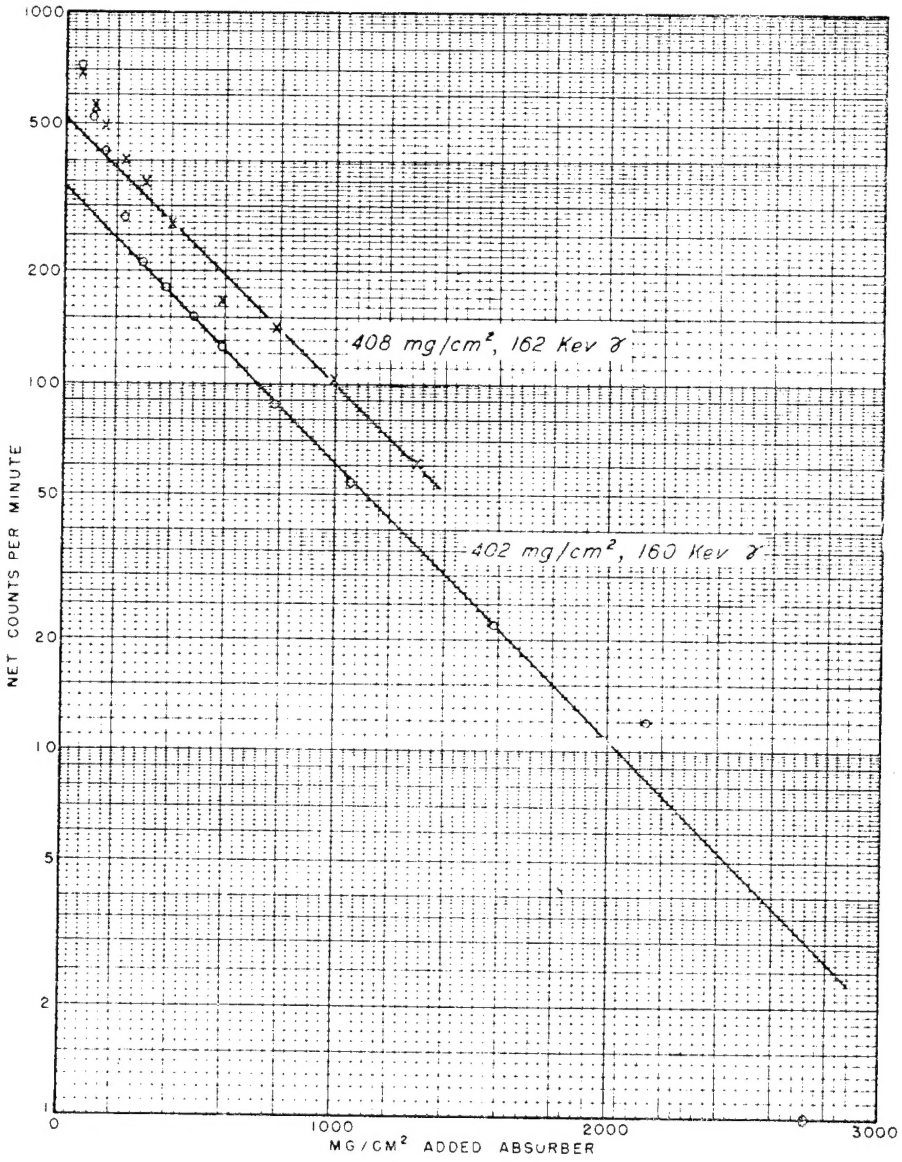

FIGURE 1

TWO Pb ABSORPTION CURVES OF U-235
BY DIFFERENCE (SEE TEXT)

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